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RE US Appl No 10/531,737 (TOS-161-USA-PCT)

### COVER MESSAGE

Please find attached hereto a 3-page Transmittal of Declaration, as well as a 5-page executed Declaration of Mr. Nakane, in the above-identified application.

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TOS-161-USA-PCT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:  
**T. Nakane, et al.**

Application Serial No.: **10/531,737**Examiner: **M.P. Chui**Filing Date: **April 18, 2005**Art Unit: **1616**Title: **Skin Treatment Composition****TRANSMITTAL OF DECLARATION**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

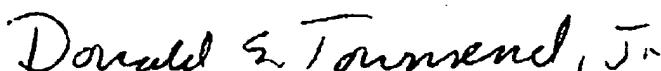
Supplemental to the Amendment filed herein on September 1, 2008, in the matter of the above-identified application, the undersigned respectfully submits herewith an executed Declaration of Mr. Nakane, said Declaration being attached hereto.

In particular, as described in the attached Declaration, a sample of antibacterial zeolite having a composition according to the present invention, as well as two samples of antibacterial zeolites not corresponding the instant invention (due to their lack of ions substitutions as claimed herein), were prepared. The color characteristics of these antibacterial zeolite compositions were then tested, exposed to the environment for 60 days, and then tested again.

The results of these tests showed that the antibacterial zeolite claimed herein provides unexpectedly superior color stability. Specifically, the test data indicated that the antibacterial zeolite of the present invention exhibits unexpectedly improved stability of the color characteristics thereof, such as hue, brightness and saturation, even under prolonged light exposure and environmental degradation.

In view of the foregoing, as well as the amendments and argument presented herein in the Amendment filed herein on September 1, 2008, it is respectfully submitted that the application is now in condition for allowance, and early action and allowance thereof is accordingly respectfully requested. In the event there is any reason why the application cannot be allowed at the present time, it is respectfully requested that the Examiner contact the undersigned at the number listed below to resolve any problems.

Respectfully submitted,



Donald E. Townsend, Jr.  
Reg. No. 43,198

Date: September 22, 2008

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CERTIFICATE OF TRANSMISSION

I hereby certify that this facsimile transmission, consisting of a 3-page Transmittal of Declaration, as well as a 5-page executed Declaration of Mr. Nakane, in U.S. patent application serial No. 10/531,737, filed on April 18, 2005, is being facsimile transmitted to the U.S. Patent and Trademark Office (Fax no. 571-273-8300) on September 22, 2008.

Donald E. Townsend, Jr.

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

**T. Nakane, et al.**

Serial No.: **10/531,737**

Art Unit: **1616**

Filed: **April 18, 2005**

Examiner: **M. P. Chui**

For: **Skin Treatment Composition**

**DECLARATION**

I, Toshihiko Nakane, do hereby depose and state the following:

1. I reside at 24-6, Tomioka-nishi 4-chome, Kanazawa-ku, Yokohama-shi, Kanagawa 236-0052 Japan.
2. I received a Bachelor's degree in Applied Chemistry from Aoyama Gakuin University in 1981. Further, I received a Master's degree in Applied Chemistry from Aoyama Gakuin University in 1983.
3. I have been employed as a researcher by Shiseido Co., Ltd., Shiseido Research Center (Shin-Yokohama), 2-1, Hayabuchi 2-chome, Tsuzuki-ku, Yokohama-shi, Kanagawa 224-8558, Japan, since April 1983. I am a co-inventor of the above-identified application.
4. The following tests, including the calculation of the color difference  $\Delta E^*$ , were conducted by me or under my direct supervision:

TOS-161-USA-PCT

**TEST PROCEDURES AND RESULTS:**

(1) Three samples #'s 1-3 of anti-bacterial zeolite powder, containing different ion compositions as shown in Table 1 below, all of which were produced by Sinanen Zeomic Co., and each having an average particle size of approximately 1.5 micrometers, and 0.5 wt% or less has a particle size over 15 micrometers, were packed up to 2 cm deep in separate glass petri dishes using vibrational packing.

(2) The brightness (L\*), hue (a\*), and saturation (b\*) values, as defined by the CIE 1976 (L\*a\*b\*) color system (as defined in the Attachment A hereto, containing an excerpt from a textbook entitled "New Cosmetic Science"), were measured for each of the three samples 1-3, using a Minolta CR-300 colorimeter.

(3) Each of the three samples of anti-bacterial zeolite prepared in (1) above was then exposed to the environment for 60 days, including exposure to daylight.

(4) At the end of the 60 day period of exposure to the environment, the brightness (L\*), hue (a\*), and saturation (b\*) values, as defined by the CIE 1976 (L\*a\*b\*) color system, were again measured for each of the three samples 1-3, using the Minolta CR-300 colorimeter mentioned above.

(5)  $\Delta E^*$ , the color difference, was then calculated for each of the three samples 1-3 using the following formula:

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}}$$

wherein L\* = brightness, a\* = hue, b\* = saturation, and  $\Delta E^*$  = change in color. The results of these calculations are shown in TABLE 1 below.

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TABLE 1

Antibacterial Zeolite Sample #	$\Delta E^*$
#1: Containing 2.5 wt % Silver, 7.5 wt% Zinc, and 2.5 % Ammonium	8.42
#2: Containing 2.5 wt% Silver, And 7.5 wt% Zinc (No Ammonium)	26.23
#3: Containing 2.5 wt% Silver (No Zinc, No Ammonium)	34.55

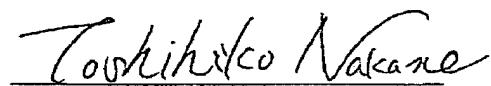
It was discovered that exposure to the environment reduces the brightness "L\*" of the powder, lowers the hue "a\*" (from white to black), and increases the saturation "b\*" (so as to cause the powder to go from white towards yellow in color).

### CONCLUSION

Based on the test data described above, and the calculated  $\Delta E^*$ , it was unexpectedly discovered that, as opposed to antibacterial zeolite powders NOT containing the ions claimed herein, an anti-bacterial zeolite containing silver, zinc and ammonium ions, as claimed in the instant application, provides exceptional color stability over time and light exposure. It is believed that this color stabilizing effect provides the presently claimed skin treatment composition with color stability and anti-staining characteristics, which are NOT provided by a composition containing an antibacterial zeolite containing silver, zinc and ammonium ions, as provided herein.

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I hereby declare that all statement made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



Mr. Toshihiko Nakane

Date: September 22, 2008

**TOS-161-USA-PCT**

**ATTACHMENT A**

# New Cosmetic Science

## Notice

The content of this book was based upon the latest information known to the authors as of the time it was written. However, information and knowledge changes constantly with the passing of time. In particular, the regulations applicable to cosmetic raw materials, pharmaceutical agents, cosmetic products and marketing activity vary considerably country by country and are subject to major revisions from time to time. Therefore, anyone who manufactures or sells cosmetic products must first investigate and confirm all applicable regulatory requirements.

Although certain examples are provided in this book of raw materials, pharmaceutical agents, formulas, etc., these are provided for purposes of general reference only in order to explain concepts of cosmetic science. No representation or guarantee of any kind is made as to their stability, safety, efficacy or the effect of present laws or other laws and regulations in the event of their actual use.

Edited by

Takeo Mitsui, Ph.D.  
Former Senior Executive Director  
and Director of Research and Development Division,  
Shiseido Co., Ltd.

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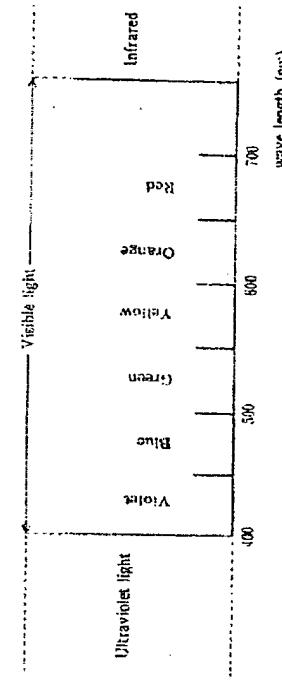


Fig. 3.1. Wavelength and color.

There is a close relationship between cosmetics and color. The search for beauty is a basic human instinct. Makeup cosmetics beautify the appearance by changing the color of the skin. In addition, color can enhance the appeal of cosmetic products which rely on color for image. The rapid developments in the science of color and in instruments for measuring color have made color management much easier, and are now utilized in the design, manufacturing and marketing of cosmetics.

Year-by-year, the regulations governing the materials used to give cosmetics their color become stricter and stricter from the safety aspect and, in each country, the use of organic color materials in cosmetics is regulated by law with regard to product quality and scope of usage. Powder materials such as inorganic pigments and extender pigments may only be used if they meet the standards of different countries for heavy metals and other impurities or are those which have received the approval of the regulatory authorities.

The technical expertise and knowledge of color scientists handling the basic constituents of cosmetics as well as accurate information related to coloring materials used in cosmetics is absolutely essential to this. Additionally, the laws related to coloring materials differ according to national legislation making it necessary to exercise caution when exporting cosmetics.

### 3 Color and cosmetic color materials

There is a close relationship between cosmetics and color. The search for beauty is a basic human instinct. Makeup cosmetics beautify the appearance by changing the color of the skin. In addition, color can enhance the appeal of cosmetic products which rely on color for image. The rapid developments in the science of color and in instruments for measuring color have made color management much easier, and are now utilized in the design, manufacturing and marketing of cosmetics.

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#### 3.1. Color

##### 3.1.1. Light and color

Light enters the eyes when they are open except when there is no light. The component of light that can be seen by the eyes is called visible light. The wavelengths longer than visible light are the infrared wavelengths and the wavelengths shorter than the visible band are the ultra-violet wavelengths. The wavelength of visible light is in the band 400–760 nm (Fig. 3.1). Natural sunlight contains many of the visible wavelengths but it is perceived by the human eye as colorless. However, when sunlight is passed through a prism, it is split into six basic colors: red, orange, yellow, green, blue, and violet.

Color is an expression of the different energy levels in light but it is not in itself a property of light. The light wavelengths are sensed by the visual receptors in the eye and the stimulation caused by each wavelength is expressed as color. In other words, color can be described as a sense.

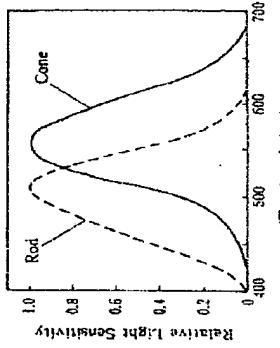


Fig. 3.2. Spectral response curve of scotopic vision (dashed line) and photopic vision (solid line).

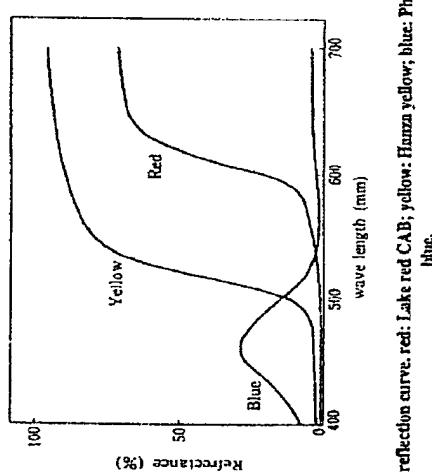


Fig. 3.3. Spectral reflection curve. red: Lake red C:AB; yellow: Hansa yellow; blue: Phthalocyanine white.

### 3.1.3. Color of coloring materials

The color of materials varies according to its composition and the type of light striking it. The same material will appear to be a different color in sunlight, fluorescent lamp and incandescent lamp. Light striking an object is either reflected off the surface of the object, back from the object interior, absorbed by the object or passed through the object. When white light strikes a colored object, the wavelength of light observed as the color is reflected and other parts are absorbed. The spectral reflection curve (Fig. 3.3) shows the reflected components of light at each wavelength in comparison with a standard object (white). The form of this curve predicts which color the object will appear. Similarly, the spectral transmission curve shows approximately which wavelengths are transmitted when white light strikes an object that transmits light. Coloring materials are chemical substances that absorb or transmit specific wavelengths. Red pigments reflect red light and absorb light other than red. In addition, red dyes absorb light other than red and transmit red light.

### 3.1.4. Three attributes of color

Color can be classified as either (1) achromatic, or colors such as white, gray, and black which neither absorb nor reflect the components of light, and (2) chromatic, or colors with hue. Chromatic colors have color as a result of absorbing part of the illuminating light or reflecting or transmitting the light in the visible light region.

Color is composed of three elements: hue, value and chroma, called the three attributes of color.

- (1) Hue: Colors such as red, yellow, green, blue and violet demonstrate this quality which is determined by wavelength.

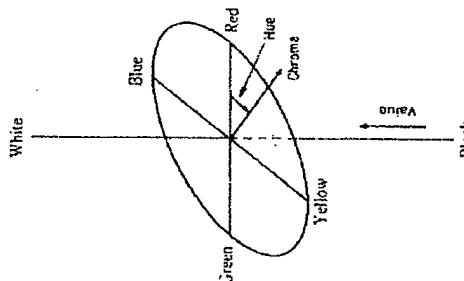


Fig. 3.4. Three-dimensional color space showing three attributes of color.

- (2) Value: The value is measured on a scale which evaluates whether the reflection from the surface of the object is high or low. When the value is high, the color is bright, and when it is low, the color is dark.
- (3) Chroma: Chroma expresses the degree of brilliance of color. A brilliant color has a high chroma value and a dull color has a low chroma value.

A diagrammatic representation of hue, value and chroma on three axes is called a three-dimensional color space (Fig. 3.4) and any color can be represented in spatial terms within this color space.

### 3.1.5. Expression of color

The human eye is able to recognize several million colors based on slight differences in the value, hue and chroma. When talking about a specific color or when recording a color, we need to be able to define that color exactly. The most accurate way is to have an actual color sample, but even in this case, there is the problem of aging-associated color changes. Ignoring color samples, for any product, it is of importance to be able to manufacture with identical color and there is a need for a systematic method of recording color as a numeric value and classifying it.

#### 3.1.5.1. Color naming methods

Trivial names: this is a system of naming colors such as salmon pink, emerald green, and lavender purple created by people who deal with colors frequently. It is called the trivial naming system. A feature of the system is that it is very similar to the original sense of color possessed by people but unless you are a specialist, it is very difficult to get an ex-

act impression of what color is meant. The system is influenced by the individual variation in people's color sense.

General color names: general color names such as red, yellowish-red, yellow, yellowish green, green, bluish green, blue, bluish violet, violet, reddish violet are adjective modifiers related to the value and chroma of color and when used with words such as light, dark, heavy, bright, etc., they can be useful in giving a more detailed impression of color. This object color naming system is described in the Japan Industrial Standards.

### 3.1.5.2. Color systems

Munsell color system: the Munsell color system is a system expressing colors in terms of hue, value and chroma. It is a system in widespread use today as it is very similar to the human color sense. In Japan, the Munsell color system has been published by the Japan Standard Association as the JIS Reference Colors<sup>3</sup>.

As shown in Fig. 3.5, hue is linked in a circle of five principal colors: red (R), yellow (Y), green (G), blue (B) and purple (P) with the addition of five intermediate colors: yellowish-red (YR), greenish-yellow (GR), bluish-green (BG), bluish-purple (BP) and reddish-purple (PR). These ten hues are divided into 10 equal parts in sensory terms and the representative hues are arranged at the 5 positions. This type of circle of colors is called a hue circle.

The value is also split into 10 equal parts in sensory terms with the achromatic colors black equal to 0, and white equal to 10.

Chroma is assigned sequential values 1, 2, 3, ... in sensory terms with achromatic colors being 0.

The Munsell color system uses this type of arrangement to express any color in terms of H/V/C. For example, 5R4/14 expresses the highest saturation of red.

CIE standard colorimetric system: in 1931, the Commission Internationale de l'Eclairage established an international system of expressing colors using tristimulus values: X, Y, and Z. In Japan, the XYZ system of describing colors is also incorporated in the JIS<sup>4</sup>.

The CIE chromaticity diagram has two axes, x and y, at a right angle on which the

spectral at each wavelength is plotted showing the spectral curve and these are joined up to obtain the spectral locus (Fig. 3.6). The chroma axis is found from the following equations.

$$x = X(X + Y + Z), \quad y = Y(X + Y + Z), \quad z = Z(X + Y + Z)$$

Since  $x + y + z = 1$ , the chromaticity can be expressed just by x and y. The projection of the line linking the achromatic point (W) and the chromatic point (S<sub>1</sub>) in the chromaticity diagram (Fig. 3.6) indicates the principal wavelength  $\lambda$  at the point (S<sub>2</sub>) where it intersects the spectral curve; the distance from W is the color "purity" (same meaning as chroma). Moreover, since y is equivalent to value, Y, x, y expresses the color.

Hunter Lab system: in 1948, R. S. Hunter proposed a color system with the ability to distinguish colors based on human senses. In this system, the tristimulus values X, Y, and

Z are converted to the equivalent L, a, and b values using the following formulae.

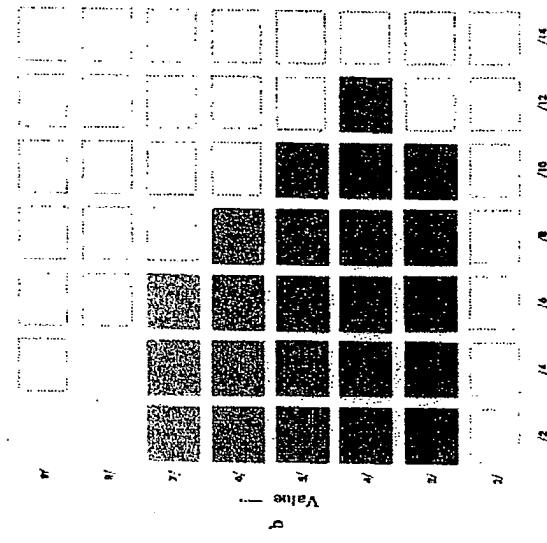
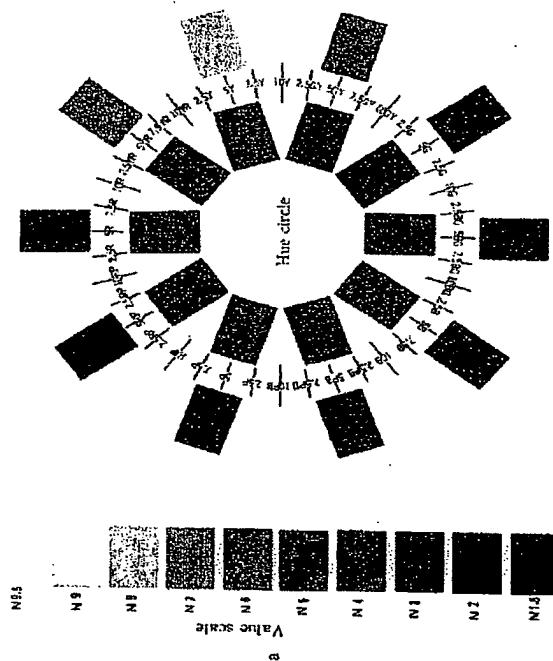


Fig. 3.5. Munsell Color System. [JIS Reference of Color Committee] JIS.

Fig. 3.6. CIE chromaticity diagram (Commission Internationale de l'Eclairage) 1931 XYZ Color System  
(Minolta Camera Corp. "Color Reading".)

$$A = 17.5(1.02 X - Y)/\Delta$$

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1000 V < 100

In addition, the color difference  $\Delta E$  between  $(L_1, a_1, b_1)$  and  $(L_2, a_2, b_2)$  can be found from the following formula.

$$\Delta E = \sqrt{1 - \frac{1}{4} \left( \frac{m_1 - m_2}{m_1 + m_2} \right)^2}$$

**L\* a\* b\*** system: the correct name for this system is the CIE 1976 ( $L^* a^* b^*$ ) Color System. This system is based on the use of tristimulus values  $X_0$ ,  $Y_0$  and  $Z_0$  correction for perfect white (reference) light reflection corresponding to  $X$ ,  $Y$  and  $Z$  of the measuring light reflection. This system is used as substitute for the Lab color system.

### 3.1.6 Color images and impression of color combinations

Every color creates various psychological impressions in the person seeing the color and these impressions vary between individuals. However, many people share the same images of different colors and understanding these images is important in designing makeup products and is an important element in the actual effectiveness of makeup.

3161 Warm and cool colors

Warm colors are colors giving a feeling of warmth and are close to red in the hue circle and include reddish-purple, red, orange, and yellow. They are the colors of the sun and fire creating an image of warmth and heat.

Coon colors are colors giving a feeling of coldness and are close to blue in the hue circle such as bluish green, blue, and bluish purple. They are colors of running water and clear lakes creating an impression of freshness, cold and intelligence.

3162 Color and emotion

Every color creates a different emotional response which may be either quite strong or relatively weak. Typical responses include pleasure, sadness, anger, security, grief and loneliness.

Table 3.1. Relationship between color and emotion

Attribute	Emotion	Color	Example of Emotion
Warm Colors	Warm, Positive Active	Red	Passion, Anger, Joy, Action, Excitement
Intermediate Colors	Yellowish Red	Yellow	Fun, Frolic, Liveliness, Health
Intermediate Colors	Green	Green	Merriment, Cheerfulness, Pleasure, Activity, Health
Cold Colors	Ordinary	Violent Green	Calmness, Relaxation, Calm, Freshness
Cool Colors	Gold	Blue	Serenity, Grace, Mystery, Anxiety, Kindness
Cool Colors	Passiveness	Blue	Restful, Coquettish, Melancholy
Cool Colors	Tranquility	Blue	Calmness and composure, Loneliness, Sorrow, Profound Tranquillity
Intermediate Colors	Bright	Blue violet	Mystery, Sublimity, Solitude
Intermediate Colors	Brightness	White	Parity, Truth
Intermediate Colors	Calmness	Gray	Calmness, Insecurity, Stemness
Intermediate Colors	Baldness	Black	Gloominess, Depression
Intermediate Colors	Dull	Black	Gloominess, Insecurity, Stemness
Intermediate Colors	Heaviness	Black	Gloominess, Insecurity, Stemness
Intermediate Colors	Freshness	Bitter orange	Ardor, Violence, Passion
Intermediate Colors	Liveliness	Bitter orange	Ardor, Violence, Passion
Intermediate Colors	Relaxation	Pink	Liveliness, Kindness
Intermediate Colors	Mildness	Pink	Liveliness, Kindness
Low	Singularity	Brown	Calmness
Low	Calmness	Brown	Calmness

Table 3.1 gives some concrete examples of which colors elicit which emotional responses<sup>6)</sup>.

### 3.1.6.3. Impression of color combinations

Although we have reported that primary colors each elicit an individual emotion, a gradient of two or more colors can elicit a variety of emotions depending on the color combination and this is extremely important in makeup coordination.

The effect of color gradation can be used to make the appearance more beautiful when used in combination with the three attributes of color, and forms the basis for the balance between contrast and coordination of color. To maintain balance, hue, value and chroma are all important, but it is essential to give consideration to hue sequence exhibited in the spectral. In the three attributes of the color system, the greatest weight is given to value contrast and hue contrast, but chroma contrast is relatively weak in comparison. Table 3.2 shows examples of the combination images of two colors.

### 3.1.7. Makeup colors

Makeup is said to be a type of art in which a number of colors are applied to the face like on a canvas. In general, women say the main purpose of using makeup is to make themselves more beautiful. Makeup is used on the bare face to create the person's ideal of beauty in harmony with other parts of the body. Consequently, makeup cosmetics are available in a variety of color tones.

Table 3.2. Examples of color combination images

Color Combination	Images
Color Combination of Same Hue	Within the same hue, a combination of different values and chromas
Combination of Similar Hues	Combination of colors in same segment of hue circle
Combination of Contrasting Hues (Complementary Hue/Contrasting hue)	Combination of Colors at Opposite Side of Hue Circle
Combination of Achromatic and Chromatic Colors	Combination of white, gray, black, and chromatic colors such as red and blue

### 3.1.7.1. Foundation colors and its applied skin color<sup>7)</sup>

The color of bare skin varies according to the race and health of the individual and also changes with season. The color of skin is mainly determined by the combined effect of the absorption of light by melanin and hemoglobin in the skin, and by transmission of light through the epidermis. For more details refer to Section 1.3. In addition, the face is different from a canvas and has location-dependent tone differences; the forehead has a relatively low value, the cheeks have a relatively high value, and under the eyes is slightly reddish.

In the case of Japan, Fig. 3.7 shows the relationship between skin color distribution of Japanese women and the foundation color distribution of the Japanese market.

The foundation color distribution is wider than the skin color distribution. Therefore, a wide variety of products are available to customers.

Since skin color changes between winter and summer, in summer it is best to use foundation colors with a lower value. In addition, a more natural three-dimensional appearance can be achieved by combining colors with slightly higher and lower values than the skin color.

The color of skin to which foundation has been applied is a mixture of the color of the bare skin and the foundation and is different from the color of the foundation meaning that care is necessary. The main cause of the difference between the color on the skin and that of the foundation is largely due to the covering effect of foundation, the application method and the amount applied.

Fig. 3.8 shows how the same skin appears to have various colors depending on the application condition (thin and thick) of an oil-based foundation; it is not a simple additive mixing of foundation color and skin color but rather skin color approaches the foundation color according to the complex curves shown in Fig. 3.8.

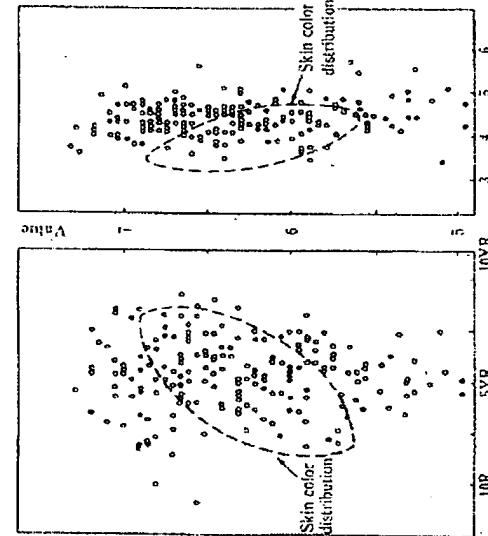


Fig. 3.7. Color distribution of commercial foundations (in Japan).

